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1. Project File No. WAG 5	· · · · · · · · · · · · · · · · · · ·	2. Project/Task _	WAG 5 Op	perable Unit 5-12	
3. Subtask <u>WAG 5 Compr</u>	ehensive RI/F	S			
4. Title: RADIUM-226 AT A	RA-01, 02, 16	i, and 23			
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RADIUM-226 AT ARA-01, 02, 16, AND 23

Radium-226 (²²⁶Ra) is a naturally occurring radionuclide in the uranium-238 (²³⁸U) decay chain found in surface and subsurface soil and rocks throughout the INEEL. The following WAG-5 sites have ²²⁸Ra listed as exceeding the risk-based concentration (RBC) of 0.5-pCi/g in soils: ARA-01 Chemical Evaporation Pond, ARA-02 Sanitary Waste Leach Field and Seepage Pit, ARA-16 Radionuclide Tank, and ARA-23 Surface Soils Around ARA-I and ARA-II. It is hypothesized that the ²²⁶Ra concentrations reported for the WAG-5 sites are representative of, and consistent with background ²²⁶Ra concentrations at the INEEL.

The author has shown in a previous EDF for the Technical Support Facility (TSF)-07 Pond at Test Area North (TAN) that reported ²²⁶Ra concentrations, as measured directly by gamma-ray spectroscopy, in soil samples are biased high (Giles 1998). The bias is associated with the analytical method used to report the concentrations, and can be accounted for and corrected with a scaling factor as determined by Giles (Giles 1998). This EDF presents a summary of the ²²⁶Ra data from the WAG-5 sites, describes the bias in the data, and presents a corrected data summary.

PROBLEM STATEMENT

²²⁶Ra at ARA-01, 02, 16, and 23

Radium-226 concentrations at the ARA facilities exceeded the RBC of 0.5-pCi/g. The concentrations reported ranged from 1.1 to 5.27-pCi/g with an average of 2.58-pCi/g. Table 1 summarizes the ²²⁶Ra soil sampling data.

Compared to the corrected, average INEEL background ²²⁶Ra concentration of 1.2-pCi/g as reported in Giles, 1998, the WAG-5 ²²⁶Ra concentrations are high. The WAG-5 ²²⁶Ra data retrieved from the Environmental Restoration Information System (ERIS) database was "flagged" with a "J." The J flag means that the result is questionable, and should be used with caution. Personal conversation with L. Don Koeppen, INEEL Sample Management Office (SMO) Radiochemist, revealed that the WAG-5 ²²⁶Ra data was flagged because the results were obtained by a direct measurement.

Table 1. Summary of WAG-5 ²²⁶Ra data.

	Concentration (pCi/g)	
Mean	2.58	
Standard Deviation	1.49	
Minimum	1.1	
Maximum	5.27	

Gamma-ray Spectroscopy Measurements for Radium-226

The use of gamma-ray spectroscopy instrumentation to measure radionuclides directly in environmental samples is a very useful tool; however, as with all tools, it must be used properly to obtain meaningful and valid results.

Soil samples contain varying quantities of naturally occurring radioactive materials (NORM). Radium-226 and uranium-235 (²³⁵U) are among the NORM constituents typically found in environmental samples. Radium-226 and ²³⁵U emit gamma-rays at energies of 186.1 and 185.7-keV, respectively. The significance of this, in terms of gamma-ray spectroscopy, is that the instrumentation does not have adequate energy resolution to distinguish between the two gamma-rays; as a result, concentrations reported for either ²²⁶Ra will be systematically biased high. The bias is quantifiable, and is presented in Attachment 1 of this EDF. (The problem with using gamma-ray spectroscopy to measure ²²⁶Ra directly is described in detail in Giles, 1998.)

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PROBLEM RESOLUTION

Scaling factors can be applied to ²²⁶Ra data; however, it must be verified that the corrections are necessary or warranted. The corrections are warranted if it can be shown that the data is representative and there is no indication that ²²⁶Ra was used in processes at ARA that resulted in releases to the environment. Additionally, it must be shown that the WAG-5 ²²⁶Ra data is representative of INEEL background. If it can be shown that the ²²⁶Ra concentrations are representative of background, then the reported values can be scaled to provide accurate numbers for input into risk assessment calculations.

- A series of steps were followed to define the nature of the ARA ²²⁶Ra data:
- Determine the analytical method used to report the ²²⁶Ra concentrations
- Evaluate the possible sources of ²²⁶Ra at the ARA facilities
- Compare the ARA data with a defined background data set
- Scale reported ARA ²²⁶Ra data to determine actual ²²⁶Ra concentrations.

The analytical method used, as previously stated, was gamma-ray spectroscopy; as a result, the ²²⁶Ra data generated is subject to special considerations. The following sections address the special considerations, and further evaluate the ARA ²²⁶Ra data.

Possible Sources of ²²⁶Ra

As mentioned before, ²²⁸Ra is a decay product in the ²³⁸U decay chain, and is present at various concentrations in nature. The primordial source of ²²⁶Ra at the ARA Sites is the ²³⁸U that is naturally occurring. The INEEL Site-wide concentration is approximately 1.0-pCi/g for ²³⁸U and its decay products, and the ²³⁸U background concentration upper tolerance limit (UTL) is 1.85-pCi/g at the 95% confidence interval (Rood et al. 1996).

Minimal process knowledge available for the ARA facilities does not provide adequate information to be able to state with any amount of certainty that 226 Ra was used in experiments or processes at the ARA facilities. If 226 Ra was used in work or process at ARA, then there is a potential that it could have been in the waste stream, or released to the environment. It may also be postulated that if 238 U or 234 U was used in any processes, 226 Ra could grow-in from their decay; however, the half lives of 238 U and 234 U, $^{4.5}$ × $^{4.5}$ × $^{4.5}$ years and $^{2.4}$ × $^{4.5}$ years, respectively, would require tens of thousands of years for the 226 Ra to grow-in to measurable quantities. As a result, the use of 226 Ra could be the only source of radium at the ARA facilities, aside from that which is naturally occurring. Statistical evaluation of the ARA data can support the hypothesis that there was no measurable 226 Ra released to the environments, and that the ARA 226 Ra concentrations are representative of background.

Statistical Evaluation of Data

The average reported ²²⁶Ra concentration for all the ARA sites is 2.58-pCi/g, which is within one standard deviation of the mean background concentration of 2.13-pCi/g reported for the background laboratory Quality Control (QC) studies of L. Don Koeppen referenced in Giles, 1998. The background laboratory QC data sets are included in Attachment 2. A Wilcoxon Rank Sum Test was performed on the ARA data to compare it to the background laboratory QC data set #1. This test was performed using the procedure presented on page 248 of Gilbert (Gilbert 1987). The test was performed to determine if the two data sets could have been drawn from the same ²²⁶Ra background population. The results of the test indicate that there is a 95% probability that the ARA data belongs to the same population as the background laboratory QC data set #1. Based on the Wilcoxon Rank Sum Test, it can be concluded that the ²²⁶Ra data is representative of INEEL background. It can also be stated that ARA processes did not release to the environment, measurable quantities of ²²⁶Ra.

Correcting the Existing WAG-5 ²²⁶Ra Data

It has been shown that in environmental samples, direct measurement of ²²⁶Ra with gamma-ray spectroscopy yields concentrations that are biased high. Scaling the reported ²²⁶Ra concentrations by 0.571 will yield correct ²²⁶Ra concentrations (Giles 1998). Table 2 shows the results of scaling the original ARA data.

Table 2. Corrected ²²⁶Ra concentrations in ARA soils (concentrations in pCi/g).

Sample ID	Reported	Corrected	Sample ID	Reported	Corrected
ARA-01 Soils			ARA-23 Soils	· 	
50103901R4	3.08	1.76	52300501R4	1.65	0.94
50104301R4	1.43	0.82	52300601R4	1.98	1.13
ARA-02 Soils			52300701R4	2.04	1.16
50200201R4	1.68	0.96	52300801R4	2.46	1.40
50200202R4	2.22	1.27	52300901R4	1.53	0.87
50200401R4	1.73	0.99	52301001R4	2.04	1.16
50200501R4	1.58	0.90	52301101R4	1.46	0.83
50200901R4	2.28	1.30	52301201R4	2.59	1.48
50201001R4	2.38	1.36	52301301R4	1.94	1.11
50201201R4	1.68	0.96	52301401R4	1.92	1.10
ARA-16 Soils			52301501R4	2.47	1.41
51600701L9	5.27	3.01	52301701R4	2.57	1.47
51600801L9	2.11	1.20	52301801R4	1.20	0.69
51600901L9	3.31	1.89	52302101R4	2.18	1.24
51601001L9	1.59	0.91	52302201R4	2.66	1.52
51601201L9	2.30	1.31	52302301R4	3.06	1.75
51601501L9	2.73	1.56	52302401R4	1.89	1.08
51601601L9	1.36	0.78	52302501R4	3.60	2.06
51601701L9	3.17	1.81	52302601R4	1.78	1.02
51602001L9	4.98	2.84	52302701R4	2.88	1.64
51602101L9	4.22	2.41	52302901R4	1.88	1.07
51602201L9	3.92	2.24	52302902R4	3.03	1.73
51602302L9	2.79	1.59	52303101R4	2.38	1.36
51602401L9	3.21	1.83	52303201R4	1.89	1.08
51602701L9	3.22	1.84	52303301R4	2.93	1.67
51602901L9	2.32	1.32	52303401R4	3.05	1.74
51603001L9	3.17	1.81	52303501R4	3.45	1.97
51603901L9	2.16	1.23	52303601R4	1.27	0.73
ARA-23 Soils			52303701R4	2.73	1.56
52300101R4	1.82	1.04	52303801R4	3.03	1.73
52300201R4	1.37	0.78	52304801R4	1.10	0.63
52300401R4	1.94	1.11			
			Mean	2.23	1.27
			Std. Dev.	0.75	0.39

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The corrected ²²⁶Ra concentrations for the ARA soils are consistent with the corrected background values, and are summarized in Table 3. The summary data in Tables 1 through 3 were prepared after the omission of an outlier, sample ID 52301901R4, with a reported ²²⁶Ra concentration of 11.9-pCi/g. This data point was more than 6-times the standard deviation away from the mean; therefore it was discarded as an outlier.

Table 3. Summary of corrected ²²⁶Ra concentrations.

	Corrected ²²⁶ Ra Concentrations (pCi/g)		
	Background #1	Background #2	ARA Facilities
Average Concentration	1.22	1.21	1.27
Minimum Concentration	0.38	0.45	0.68
Maximum Concentration	2.59	2.20	3.01

In-Situ Gamma-Ray Spectroscopy Measurements

In-situ gamma-ray spectroscopy measurements were made in two deep boreholes that were drilled in the ARA-01 pond area. The purpose of the measurements was to determine the vertical extent of man-made radionuclide migration in the pond subsurface, and the results are summarized in Section 3.1.1 of this document. Although the man-made contamination was minimal, and confined to the surficial sediments, the in-situ gamma-ray measurements also provided valuable information with regards to the NORM in the subsurface materials. Concentrations of ²²⁸Ra daughters were measured throughout the two boreholes. The daughter products, bismuth-214 (²¹⁴Bi) and lead-214 (²¹⁴Pb), are found in a state of secular equilibrium with their parent, ²²⁸Ra, in INEEL soils and rocks. This condition means that the concentrations of the daughter products and the parent are equal; therefore, if the activity of one of the radionuclides in a decay chain can be measured, then the same activity can be assigned to the other radionuclides in that chain. Radium daughters are used to indirectly quantify ²²⁸Ra in soil samples using gamma-ray spectroscopy, because the gamma-rays emitted by ²¹⁴Bi and ²¹⁴Pb produce "clean" peaks in the gamma-ray spectra, without interference problems from other radionuclides. The in-situ data is summarized below in Table 4.

Table 4. In-situ ²¹⁴Bi and ²¹⁴Pb data summary for ARA-01.

	Assigned ²²⁶ Ra Concentrations (pCi/g)		
	Borehole #1	Borehole #2	
Average Concentration	0.28	0.64	
Standard Deviation	0.15	0.18	
Minimum Concentration	0.13	0.37	
Maximum Concentration	0.71	0.96	

The ²²⁶Ra concentrations appear lower in the boreholes with respect to the ARA soils because the boreholes were drilled into the basalt. This is consistent with other in-situ gamma-ray measurements made at other facilities at the INEEL in that the basalt has much lower natural radioactivity than the surficial soils. This observation at the ARA-01 pond further supports the argument that the ²²⁶Ra at this site is at background.

CONCLUSION

This EDF provides a strong, valid argument that the ²²⁶Ra concentrations in the ARA-01, 02, 16, and 23 soils, and the ARA-01 subsurface basalt are at background levels. The statistical comparison concludes with 95% confidence, that the ²²⁶Ra concentrations are representative of, and consistent with INEEL background values.

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The ²²⁶Ra data collected during FY1997 WAG-5 Comprehensive Remedial Investigation/Feasibility Study (RI/FS) are at background levels. This has been validated statistically and through comparison with other data sets. The previously mentioned WAG-5 sites should not be held for further actions based on the ²²⁶Ra concentrations.

RECOMMENDATION

The direct measurement of ²²⁶Ra in environmental samples using gamma-ray spectroscopy yields concentrations that are systematically biased high. The ARA data is the second data set from the INEEL ERIS database that has been reviewed for this bias, and for the second time, it has been concluded that the ²²⁶Ra data was representative of background. This will probably not be the last time ²²⁶Ra will be identified incorrectly as posing an unnecessary risk at the INEEL. The primary reason is that the INEEL background ²²⁶Ra concentrations (~1-pCi/g) exceed the RBC of 0.5-pCi/g. This is compounded by the reporting of ²²⁶Ra concentrations that are systematically biased high due to direct measurement with gamma-ray spectroscopy. This problem with the ²²⁶Ra concentrations needs to be addressed to eliminate the confusion in the future, and more importantly, to eliminate unnecessary and costly remedial actions. Primary tasks that should be completed are as follows:

- Remove ²²⁶Ra from the gamma-ray spectroscopy target list (QAPjP for WAGs 1-7 & 10, and the SMO Master Task Subcontract Statement of Work)
- Review all ²²⁶Ra in soils data from the ERIS database, and determine that which needs correction
- Perform a Site-wide study to establish the INEEL background for ²²⁶Ra using a combination of laboratory and in-situ measurements
- Compare existing data to the established background to determine which facilities have a true contamination problem with ²²⁶Ra
- Perform future ²²⁶Ra measurements by other than direct measurement of the 186.1-keV gamma-ray.

The first task is currently being completed with the revision of the SMO SOW. The other tasks are recommended to eliminate the biased ²²⁶Ra data in the ERIS database, and also to establish a well defined ²²⁶Ra background for the INEEL soils.

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ATTACHMENT 1

Method for Removing the ²³⁵U Contribution to the ²²⁶Ra Reported Concentrations as Determined Directly by Gamma-Ray Spectroscopy

Attachment 1

Method for Removing the 235U Contribution to the ²²⁶Ra Reported Concentrations as Determined Directly by Gamma-Ray Spectroscopy

Direct measurement of ²²⁶Ra by gamma-ray spectroscopy in environmental samples is often difficult, and consistently reports concentrations that are inaccurately high. The reason for the inaccuracy is failure to correct the data for the interference from ²³⁵U. Both ²²⁶Ra and ²³⁵U are present in soils naturally. Uranium is the parent radionuclide in the Actinium decay series, and ²²⁶Ra is one of the progeny in the ²³⁸U decay chain. Although members of different decay series, both ²²⁶Ra and ²³⁵U decay by alpha particle emission, followed by the emission of gamma-rays. The interference in the ²²⁶Ra measurement comes from the 185.739-keV gamma-ray emitted by ²³⁵U. This is a problem because ²²⁶Ra emits a gamma-ray at 186.11-keV (Browne & Firestone), and for the high-purity germanium (HPGe) detectors used for analysis, these gamma-rays are indistinguishable.

The following derivation is a method that can be used to resolve the individual contributions of ²³⁵U and ²²⁶Ra to the composite peak at 186-keV in a gamma-ray spectrum.

GIVEN

Energies and Branching Ratios of Gamma-rays (Browne & Firestone 1986):

$$E_{U^{235}} = 185.739 keV$$
 $N_{U^{235}} = 0.53 \frac{\gamma}{dis}$

$$E_{Ra^{226}} = 186.11 keV$$
 $N_{Ra^{226}} = 0.0328 \frac{\gamma}{dis}$.

Isotopic Abundance's of Uranium Isotopes in soils (Browne & Firestone 1986):

$$I_{U^{235}} = 0.720\%$$

$$I_{U^{238}} = 99.2745\%$$

Specific Activities of Uranium Isotopes in natural uranium (Eisenbud 1987):

$$SA_{U^{235}} = 1.54 \times 10^4 \frac{pCi}{g} (^{nat}U)$$

$$SA_{U^{238}} = 3.33 \times 10^5 \frac{pCi}{g} (^{nat}U)$$

The ratio of the Specific Activities is then:

$$\frac{SA_{U^{235}}}{SA_{U^{238}}} = \frac{1.54 \times 10^4 (pCi/g)}{3.33 \times 10^5 (pCi/g)} = 4.667 \times 10^{-2}$$
(1-1)

ASSUMPTIONS

• It is assumed that the ²³⁸U decay chain is in secular equilibrium in the sample.

This assumption is based on the fact that the ²³⁸U equilibrium is well established in the INEEL soils (Rood et al. 1996), and that the equilibrium is undisturbed during the sample preparation and preservation (McHugh). It is re-emphasized here that the samples and results from the L. Don Koeppen and the 1989 TSF-07 pond data sets were collected in sealed containers, and counted with no sample preparation.

• The counting efficiencies of any given HPGe detector system is nearly identical for the two gamma-ray energies in question (i.e. 185.739-keV, and 186.11-keV).

It can be shown that the efficiencies are identical, or nearly so, by the following:

The absolute counting efficiency, $\varepsilon(E)$, of a detector system can be defined in very general terms as:

$$\varepsilon(E) = k \cdot E \tag{1-2}$$

where:

E = gamma-ray energy in keV

k = empirically determined constant.

Furthermore, for any given detector system, k is a constant for all energies such that

$$k = \frac{\varepsilon(E_1)}{E_1} = \frac{\varepsilon(E_2)}{E_2} \tag{1-3}$$

$$\frac{\varepsilon(E_1)}{\varepsilon(E_2)} = \frac{E_1}{E_2} \tag{1-4}$$

Specifically,

$$\frac{\varepsilon(185.739)}{\varepsilon(186.11)} = \frac{185.739}{186.11} = 0.998 \tag{1-5}$$

This shows that the difference in the efficiencies is less than 0.2%, and does not significantly contribute to the remainder of the calculations presented in this paper.

CORRECTION FACTOR DERIVATION

If we have a 1-gram sample of soil, and it has been determined that the ²³⁸U concentration is 1-pCi/g, then from Equation 1 above, the ²³⁵U concentration is 4.667x10⁻²-pCi/g. Furthermore, the respective total activities in the 1-g sample are 1-pCi and 4.667x10⁻²-pCi for ²³⁸U and ²³⁵U. Using the assumption that the ²³⁸U decay chain is in equilibrium, then we can also assume the ²²⁶Ra concentration in the sample is 1-pCi. In terms of Bequerels (disintigrations/second), these activities are:

$$A_{Ra^{226}} = 3.7 \times 10^{-2} Bq$$
$$A_{U^{235}} = 1.727 \times 10^{-3} Bq$$

These values can be used to calculate the gamma-ray production rates from ²²⁶Ra and ²³⁵U in our sample:

$$\gamma_{Ra} = (3.7 \times 10^{-2} \, dis/s) \cdot (0.0328 \gamma \, / dis) = 1.21 \times 10^{-3} \, \gamma \, / s \tag{1-6}$$

$$\gamma_U = (1.727 \times 10^{-3} \, dis/s) \cdot (0.53\gamma \, / dis) = 9.15 \times 10^{-4} \, \gamma \, / s \tag{1-7}$$

To quantify the individual contributions of ²²⁶Ra and ²³⁵U to the 186-keV gamma-ray signal, we simply take the ratio of the gamma-ray production rates:

$$\frac{\gamma_{Ra^{226}}}{\gamma_{U^{235}}} = \frac{1.21 \times 10^{-3} \, \gamma \, / s}{9.15 \times 10^{-4} \, \gamma \, / s} = 1.33 \tag{1-8}$$

In fractional form, this can be written as:

$$\frac{\gamma_{Ra^{226}}}{\gamma_{U^{235}}} = \frac{4\gamma/s}{3\gamma/s} \tag{1-9}$$

This means that for every seven gamma-rays that are counted by the detector system (at 186-keV), four gamma-rays are from ²²⁶Ra, and three gamma-rays are from ²³⁵U. In terms of a decimal equivalency, 57.1% of the gamma-rays are from ²²⁶Ra, and 42.9% gamma-rays are from ²³⁵U.

The relationship developed in Equation 9 can be used to determine the actual ²²⁶Ra concentration from the reported concentration. Simply multiplying the reported concentration by 0.571 will yield the correct ²²⁶Ra concentration.